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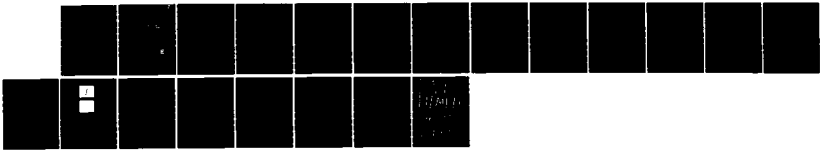

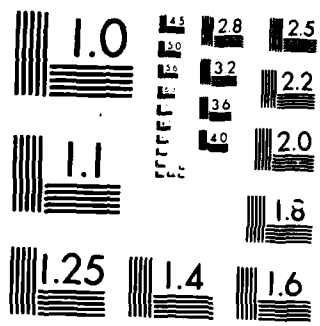

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Technical Report
737

Effect of Magnetostriction on Remanent Magnetization

G.F. Dionne

5 December 1985

Lincoln Laboratory

MASSACHUSETTS INSTITUTE OF TECHNOLOGY

LEXINGTON, MASSACHUSETTS

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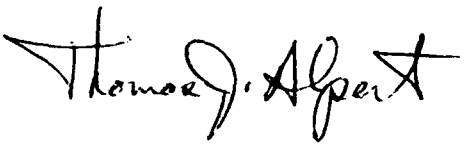

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EFFECT OF MAGNETOSTRICTION ON REMANENT MAGNETIZATION

G.F. DIONNE

Group 33

TECHNICAL REPORT 737

5 DECEMBER 1985

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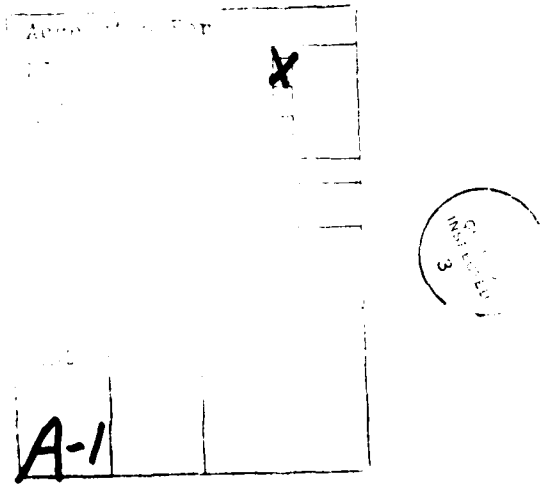
LEXINGTON MASSACHUSETTS

ABSTRACT

Magnetostriction effects on remanent magnetization are examined in a phenomenological model that is used to design a high magnetization polycrystalline spinel ferrite with excellent square hysteresis loop properties. Isotropic magnetostriction is compensated by substituting Mn^{3+} ions for 0.2 Fe^{3+} ions per formula unit of $Ni_{.65}Zn_{.35}Fe_2O_4$ host material. As predicted by theory, the magnetostriction reduction dramatically increases the remanent magnetization and removes most of its external stress sensitivity. The resulting remanent magnetization, which approaches 4000 G, represents an increase of more than a factor of 2 over that of the host composition, with no significant deterioration in other magnetic and dielectric properties. The remanence ratio improvement is in general accord with estimates predicted by the theoretical model.



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EFFECT OF MAGNETOSTRICTION ON REMANENT MAGNETIZATION

1. INTRODUCTION

Polycrystalline ferrite materials with square hysteresis loops can be difficult to obtain for a number of reasons. The demagnetizing effects that determine the magnetization in the remanent state (i.e., zero applied magnetic field) arise from averaged magnetocrystalline anisotropy of randomly oriented grains, closure and reverse domains at pores and grain boundaries, and magnetostriction which can in turn influence both anisotropy and domain wall energies. In this report, the phenomenon of demagnetization from magnetostrictive strains will be examined.

A comprehensive discussion of the effects of magnetostriction on the creation of reverse domains was published by Goodenough.[1] This theory helped to explain the use of external stress in compensating magnetostriction effects to achieve square loops in memory cores. The effect of uniaxial stress on the magnetocrystalline anisotropy was later analyzed by Dionne,[2,3] who drew conclusions similar to Goodenough, but also placed the concept on a more quantitative basis by a demonstration of how magnetostriction compensation through Mn^{3+} additions could produce stress-insensitive iron garnet compositions. A further result of this work was the discovery that the amount of Mn^{3+} required for cancellation of external stress effects also produced a maximum remanent magnetization,[4] confirming to some extent the predictions of the Goodenough theory.

In this present work, we shall introduce a simple theoretical concept to explain magnetostrictive demagnetization, and then apply its results to analyze the characteristics of a high magnetization NiZn spinel ferrite with square hysteresis loop properties.

2. THEORETICAL CONSIDERATIONS

A. Phenomenological Model

For isotropic (polycrystalline) materials, magnetostriction phenomena feature hysteresis behavior[5] similar to the magnetization process (see Figure 1). This result is not unexpected because of the cause-and-effect relation between magnetocrystalline anisotropy and lattice stress. As the applied field H rotates the magnetization M into a hard direction, the attendant stress produces a magnetostriction strain λ of energy $(1/2)\ C\lambda^2$ in the direction of H, where C is the Young's Modulus. As a consequence, this magnetoelastic coupling produces a monotonic relation between M and λ components in the direction of H. With the magnetic field removed, M_r and λ_r represent the remanent magnetization and magnetostriction.

Consider as a general assumption that $M = M_s\ f(H)$ and $\lambda = \lambda_s\ g(H)$, where the subscript s refers to the saturation values. If we also assume for convenience that $g(H) = [f(H)]^n$, where $n > 0$, it follows that

$$\lambda = \lambda_s\ (M\ M_s)^n \quad . \tag{1}$$

At remanence, the material is in a state of strain with energy $(1/2)\ C\lambda_r^2$.

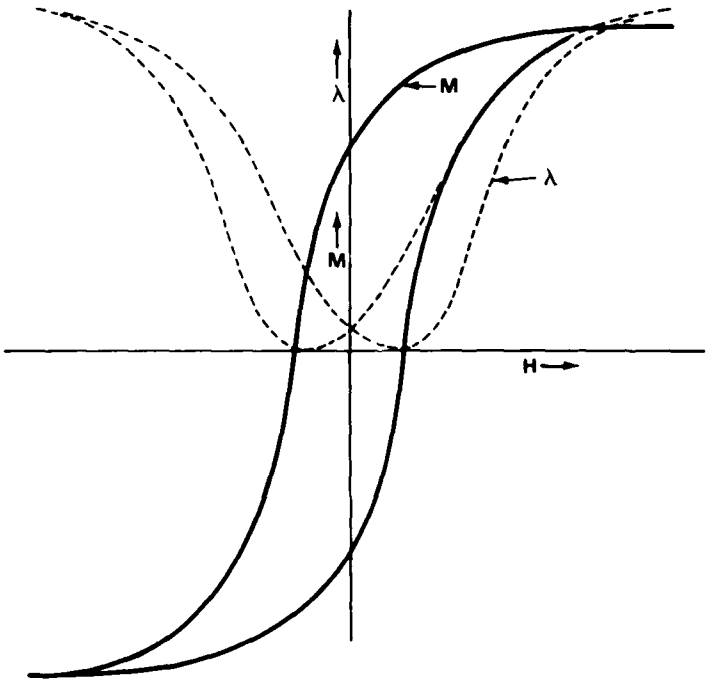

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Figure 1. Hysteresis effects of magnetization and magnetostriction.

As depicted in Figure 2, the internal stress causing the strain may be relieved if part of the magnetization changes its direction, thereby reducing M_r and establishing a new energy equilibrium. If the coercive field H_c is treated as a bias field in the direction of H,* then it follows that the internal stress must produce its own effective field to overcome the coercive field H_c and create partial switching or demagnetizing domains. If we express the system energy density as the sum of strain and magnetic contributions,

$$E = (1/2)\, C\lambda^2 - MH_{eff} \quad , \tag{2}$$

or, with substitution for λ from Equation (1)

$$E = (1/2)\, C\lambda_s^2\, (M/M_s)^{2n} - MH_{eff} \quad , \tag{3}$$

where $H_{eff} = H + H_c$. After minimizing E with respect to M (i.e., $dE/dM = 0$) we find that the equilibrium magnetization is determined by

$$M/M_s = (H_{eff}M_s/nC\lambda_s^2)^{1/(2n-1)} \quad . \tag{4}$$

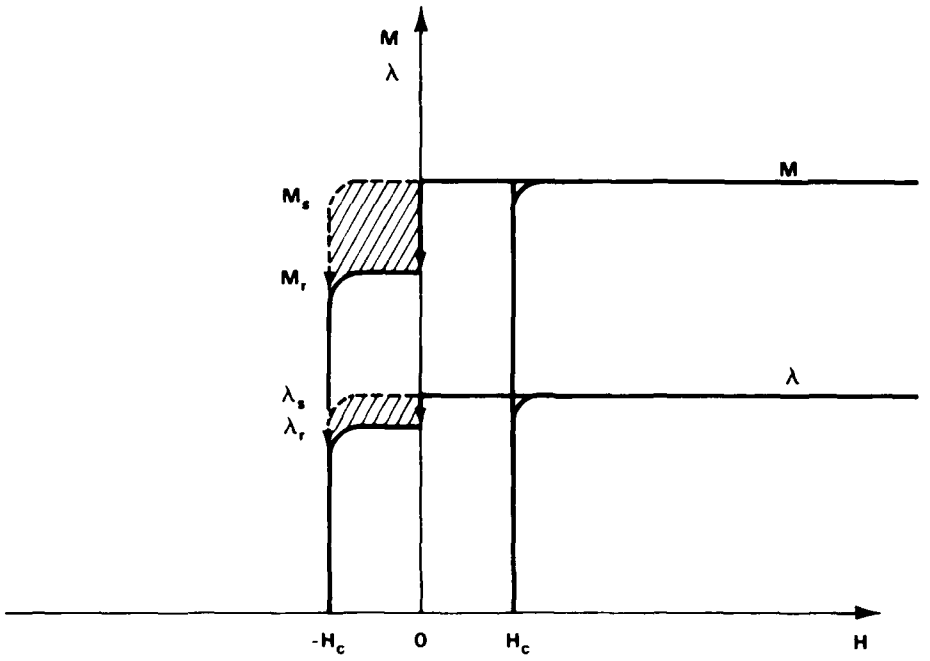

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Figure 2. Reduction of remanent magnetization M_r by magnetostrictive demagnetization.

* The sign of H_c in this context would always be in the sense that opposes domain wall motion. Thus, if the applied field were increasing toward saturation, H_c would be of opposite sign; if H were becoming smaller, as it approaches remanence in this case, H_c would reinforce it. This definition is used in the construction of the curves in Figure 6.

For the specific case of H = 0 (i.e., at remanence), Equation (4) is reduced to:

$$R = M_r / M_s = (R_1/n)^{1/(2n-1)} \quad \text{for } 0 \leqslant R \leqslant 1 \quad , \tag{5}$$

where $R_1 = H_c M_s / C\lambda_s^2$. In Figure 3, R is plotted as a function of R_1 for n = 1, 2, and 3.

Two comments are appropriate here. First, the direct dependence of R on H_c points to a higher degree of magnetostrictive demagnetization where the coercive field is small. Second, the inverse dependence of R on λ_s^2 indicates not only its high sensitivity to magnetostriction, but also its independence of the sign of λ_s. Unlike the external stress effect which can cause R to increase or decrease according to the relation between the respective signs of λ_s and external stress σ (Reference 3), i.e., whether the applied stress reinforces or opposes the internal stress, the magnetostrictive strain caused by internal stress will always decrease R in proportion to λ_s^2.

An additional effect of magnetostriction can be a decrease in H_c. According to Lee,[6] the magnetocrystalline anisotropy $K_1 = K_0 + \Delta K$, where K_0 and ΔK are the unstrained and magnetostrictive contributions, respectively. For a crystal of cubic symmetry

$$\Delta K = (9/4)[(c_{11} - c_{12})\lambda_{100}^2 - 2c_{44}\lambda_{111}^2] \quad , \tag{6}$$

where λ_{100} and λ_{111} are the respective magnetostriction constants along [100] and [111] axes, and c_{ij} represents the elastic constants. Since the strain energy occurs as a result of stress caused by

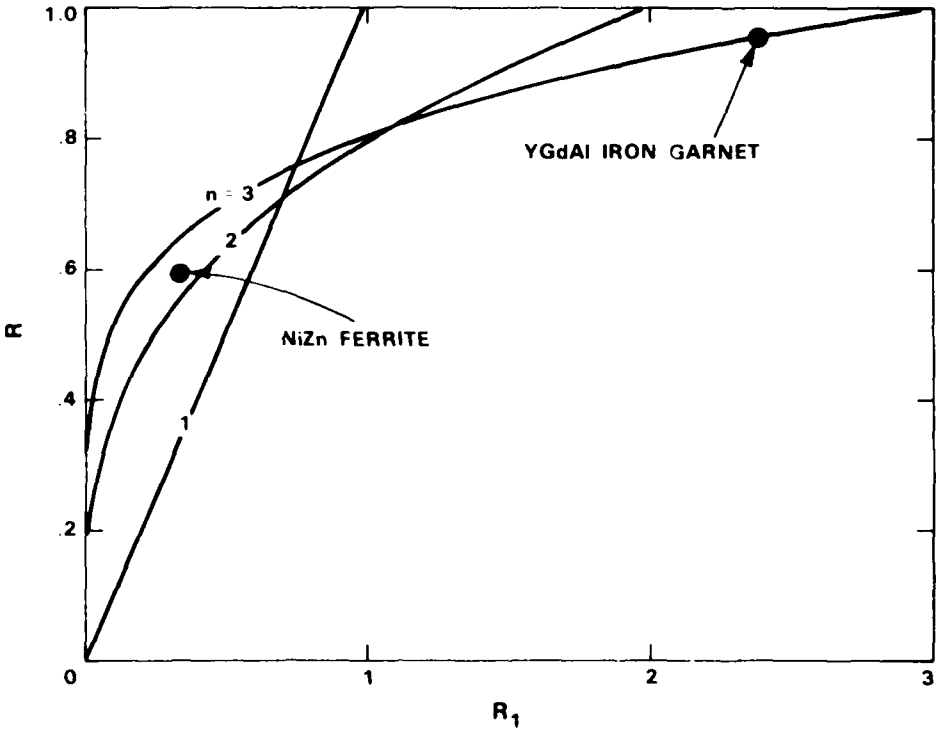

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Figure 3. R versus R_1 for n = 1, 2, and 3.

anisotropy, it is expected that the net anisotropy would be reduced and that K_0 and ΔK would be of opposite sign.

A decrease in the magnitude of K_1 would not only facilitate the demagnetization effect discussed above by lowering domain wall energies to reduce H_c, but would also raise the initial permeability. Magnetostriction is at least partly responsible for the correlation between low remanent magnetization and high initial permeability.

B. Chemical Composition

To test the above model with a spinel ferrite, a commercial nickel-zinc ferrite* closely based on the composition $Ni_{.65}Zn_{.35}Fe_2O_4$ was chosen as the reference host because of its low remanence ratio and coercive field, and high magnetostriction.

Applying the general relation for the isotropic magnetostriction constant for a cubic lattice, $\lambda_s = (2/5)\lambda_{100} + (3/5)\lambda_{111}$, we calculate room temperature λ_s values from the data in Table I for $NiFe_2O_4$ (Reference 7) as -31×10^{-6} and $MgFe_2O_4$ (Reference 8) as -3×10^{-6}. Since the Fe^{3+} content is the same for both, we may use the single-ion approximation to estimate the Ni^{2+} contribution as $\lambda_s(Ni) = \lambda_s(NiFe_2) - \lambda_s(Fe_2) = -28 \times 10^{-6}$. For the $Ni_{.65}Zn_{.35}Fe_2O_4$ composition, $\lambda_s = 0.65\lambda_s(Ni) + \lambda_s(Fe_2) = -21 \times 10^{-6}$. Since Mn^{3+} ions have been used to compensate λ_s in both $MgFe_2O_4$ (Reference 8) and $Li_{.5}Fe_{1.5}O_4$ (Reference 9), it is logical that Mn^{3+} ions should have similar effects in the NiZn ferrite family.

TABLE I

Magnetostriction Constant Values (300 K)

Composition	λ_{100} ($\cdot 10^{-6}$)	λ_{111} ($\cdot 10^{-6}$)	λ_S ($\cdot 10^{-6}$)	Reference
$NiFe_2O_4$	-45.9	21.6	-31	7
$MgFe_2O_4$	-11.1	+2.3	-3	8
$Ni_{.65}Zn_{.35}Fe_2O_4$	-33.7	13.2	-21	estimate
$Y_3Fe_5O_{12}$	-1.3	2.7	-2.1	3
$Y_3Fe_{4.74}Mn_{0.26}O_{12}$	+16.2	+1.1	+7.1	3

* Catalog No. TT 2-111, Trans-Tech, Inc.

Based on the results of the successful λ_s compensation in $Y_3Fe_5O_{12}$ (YIG), the single-ion λ_s contribution of octahedral-site Mn^{3+} ions as postulated in the local-site distortion model reported earlier[10] is expected to be about $+36 \times 10^{-6}$ per Mn^{3+} ion per formula unit in the garnet lattice. If this quantity is scaled to the spinel lattice where there are 3 8 fewer cations per formula unit, its value increases (by 8 3) to $+95 \times 10^{-6}$. Pursuing this reasoning one step further, we find that magnetostriction compensation ($\lambda_s = 0$) should occur with about 0.2 Mn^{3+} ions per formula unit to balance the 21×10^{-6} value of the host material. As a consequence, the composition chosen was $Fe^{3+}_{.65}Zn^{2+}_{.35}[Ni^{2+}_{.65}Fe^{3+}_{1.15}Mn^{3+}_{.2}]O_4$

3. EXPERIMENTAL RESULTS

The test specimens were cut from sintered bars of high density prepared by conventional ceramic techniques. In Figure 4 the room temperature hysteresis loops of the NiZn and NiZnMn ferrites prepared under identical conditions are compared to demonstrate the dramatic effect of λ_s compensation by Mn^{3+} ion substitutions. The remanent magnetization $4\pi M_r$ increased by a factor of 2 to approach 4000 G, with only a small increase in coercive field. Verification of the near elimination of magnetostriction was established by the sharp decrease in the sensitivity of $4\pi M_r$ to a uniaxial external compressive stress (see Figure 5). Values of other parameters are presented in Table II, together with earlier unpublished data on remanence ratio enhancement with Mn^{3+} substitution into an YGdAl iron garnet composition.

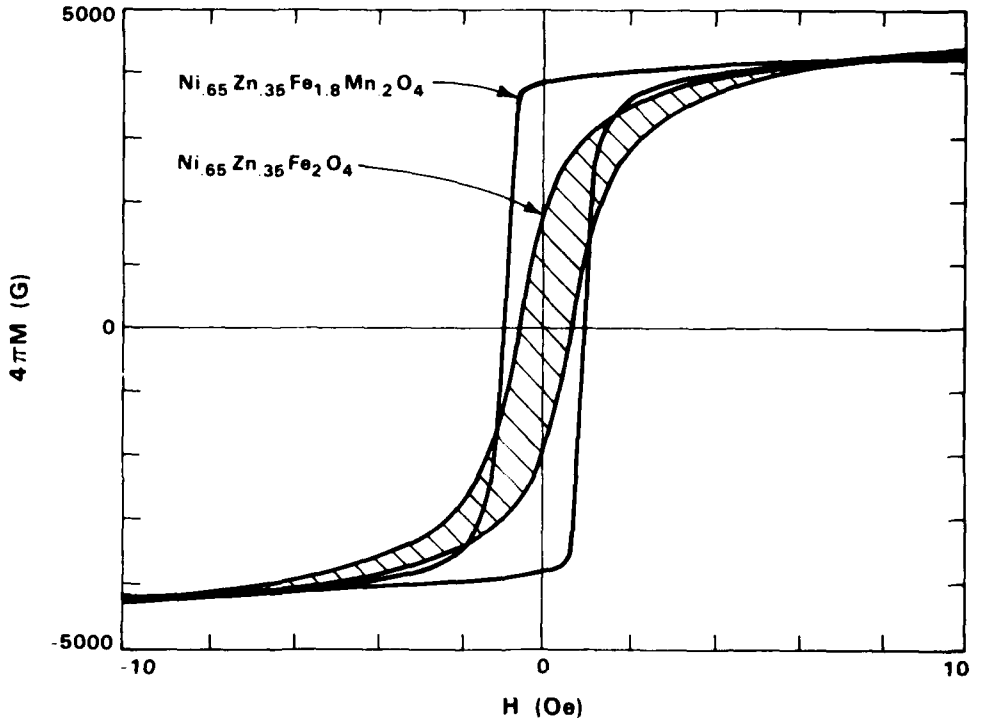

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Figure 4. Comparison of low field hysteresis loops of $Ni_{.65}Zn_{.35}Fe_2O_4$ and $Ni_{.65}Zn_{.35}Fe_{1.8}Mn_{.2}O_4$

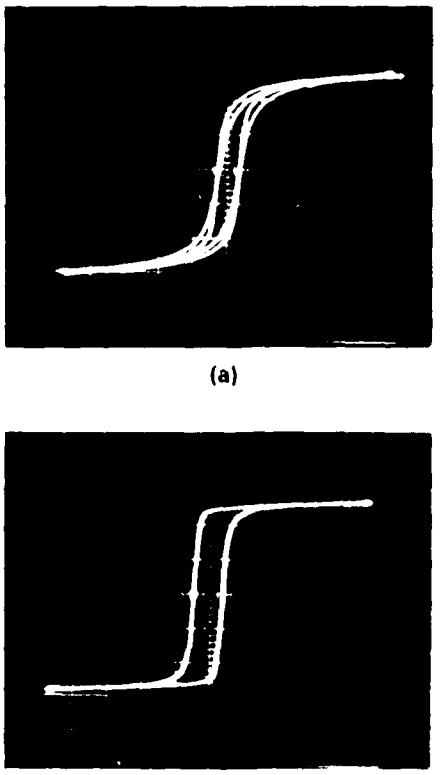

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Figure 5. External stress sensitivity of (a) $Ni_{.65}Zn_{.35}Fe_2O_4$ and (b) $Ni_{.65}Zn_{.35}Fe_{1.8}Mn_{.2}O_4$. Compressive stress ($\sim 5 \times 10^7$ dyn-cm^{-2} in this case) applied parallel to the magnetic field normally increases the remanent magnetization in these materials.

If we assume that $C \simeq 30 \times 10^{11}$ dyn-cm^{-2} and $H_c \simeq 1$ Oe for both NiZn ferrite (M_s = 400 G) and the iron garnet composition (M_s = 64 G) in Table II, and that the respective uncompensated λ_s values are approximately -21×10^{-6} and 3×10^{-6}, the R_1 value would be 0.36 for the NiZn ferrite and 2.4 for the YGdAl iron garnet. From the curves of Figure 3, the R decreases from magnetostrictive demagnetization in each case could be explained by a value of n in the 2 to 3 range.

In Figure 6, Equation (4) is plotted as a function of H applied for R_1 = 0.36 and n = 2.5 in an attempt to simulate the magnetostrictive effects on the low field hysteresis loop. The discrepancy between the calculated and measured curves shows the influence of anisotropy and microstructural demagnetization which were not taken into account by the simple theoretical model.

TABLE II

Magnetic and Dielectric Data (300 K)

Parameter	NiZn Spinel	NiZnMn Spinel	YGdAl Garnet	YGdAlMn Garnet
$4\pi M_s$ (G)	4960	4697	775	747
$4\pi M_r$ (G)	1900	3819	480	508
R	0.38	0.81	0.62	0.68
ΔR	—	+0.43	—	+0.06
H_c (Oe)	0.7	1.0	0.9	1.4
Dielectric Constant	12.38	12.48	14.82	14.91
Dielectric Loss Tangent at 9 GHz	$<10^{-3}$	$<10^{-3}$	$<2\times10^{-4}$	$<2\times10^{-4}$

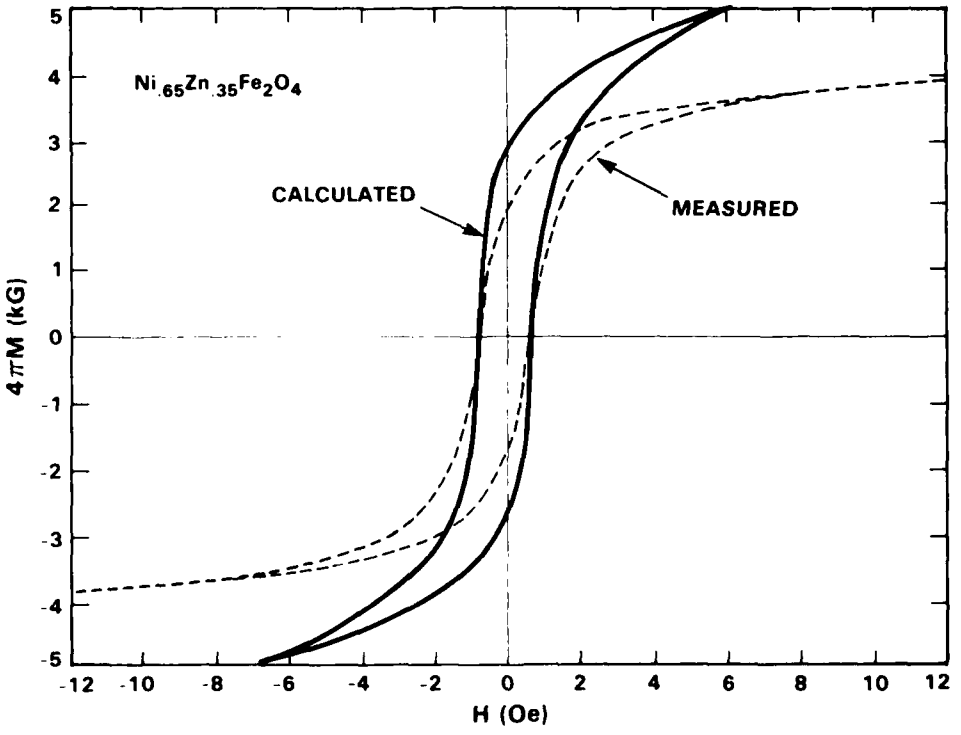

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Figure 6. Comparison of measured loop of $Ni_{.65}Zn_{.35}Fe_2O_4$ with calculated estimate based on Equation (4). The sign of H_c is always chosen to oppose domain wall motion.

The observed change in H_c with the substitution of Mn^{3+} ions is consistent with the prediction of Equation (6). If the increase in H_c is attributed to the effective elmination of magnetostriction, then ΔK for the host composition must be of oppostie sign to K_1 and of significant magnitude. To make this computation, we use the λ_{100} and λ_{111} estimates listed in Table I and the elastic constants for $MgAl_2O_4$ spinel $c_{11} = 30.0 \times 10^{11}$, $c_{12} = 15.2 \times 10^{11}$, and $c_{44} = 15.9 \times 10^{11}$ dyn-cm^{-2} (Reference 11). In this manner, ΔK is estimated to be greater than $+10^3$ ergs-cm^{-3}, as compared with a typical K_1 value of the order of -10^4 ergs-cm^{-3}.

4. SUMMARY

Enhancement of remanent magnetization in spinel and garnet ferrites, particularly those with low coercive fields, may be achieved through reduction of magnetostriction. The phenomenon of stress demagnetization may be interpreted as a decrease in energy by partial switching of the magnetization direction to relieve magnetostrictive strains in the remanent state. Cancellation of the isotropic magnetostriction constant may thus remove the internal stress demagnetization, as well as the sensitivity to external stress. The predictions of this simple theoretical model were verified by the dramatic increase in the remanent magnetization when a designed concentration of Mn^{3+} ions was used to reduce magnetostriction in a high magnetization NiZn spinel ferrite composition. Complete magnetostriction compensation in this NiZn ferrite family appears to be possible by composition refinements currently in progress.

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Magnetostriction effects on remanent magnetization are examined in a phenomenological model that is used to design a high magnetization polycrystalline spinel ferrite with excellent square hysteresis loop properties. Isotropic magnetostriction is compensated by substituting Mn^{3+} ions for 0.2 Fe^{3+} ions per formula unit of $Ni_{.65}Zn_{.35}Fe_2O_4$ host material. As predicted by theory, the magnetostriction reduction dramatically increases the remanent magnetization and removes most of its external stress sensitivity. The resulting remanent magnetization, which approaches 4000 G, represents an increase of more than a factor of 2 over that of the host composition, with no significant deterioration in other magnetic and dielectric properties. The remanence ratio improvement is in general accord with estimates predicted by the theoretical model.

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